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journal or publication title	CYRIC annual report
volume	1980
page range	131-133
year	1980
URL	http://hdl.handle.net/10097/48589

V. 25 Mössbauer Effect Measurements on Valence Fluctuating Compound Sm_3Se_4

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Magnetic susceptibility^{1,2)}, transport properties³⁾, specific heat²⁾, lattice constant²⁾ and X-ray L_{III} absorption⁴⁾ measurements of Sm_3Se_4 reported so far are similar to those of Eu_3S_4 and Sm_3S_4 , indicating the thermal activated valence fluctuation.⁵⁾ Ac-conductivity data²⁾, however, seem to suggest a quantum-mechanical tunnelling valence fluctuation at lowest temperature of measurement different from the sulphides. The aim of the present work is to study the electronic configuration of Sm_3Se_4 at various temperatures by Mössbauer measurements, in which the isomer shift should provide a data for accurate determination of the 4f configurations in Sm ions. The 4f⁵ and 4f⁶ configurations produce different potentials on the various s electrons, which produce a change of the amplitude of these s electrons on the nucleus and thus give rise to different isomer shifts for the two configurations.

The Mössbauer absorption spectra were obtained in a standard transmission geometry, using a conventional constant-acceleration spectrometer. The temperature of the source and that of the absorber could be changed independently from 4.2 K up to room temperature. Measurements were performed by using the 22.5 keV γ -transition in ^{149}Eu . The γ -rays were detected by a pure Ge LEPS (Low-Energy Photon Spectrometer) of 5 cc. The sample velocity was calibrated by detecting the laser light reflected from a mirror attached to the rear end of the source driver. The source consisting of ^{149}Eu was produced by the (p, 2n) reaction using the cyclotron of Tohoku University. Target was a pressed pellet of $^{150}\text{Sm}_2\text{O}_3$ containing 280 mg of enriched ^{150}Sm isotope. The absorber consisted of a polycrystalline sample of Sm_3Se_4 produced from a pure single crystal containing ^{149}Sm in natural abundance. The same single crystal had been used for previous experiments performed in our laboratory.²⁾

Mössbauer spectra of Sm_3Se_4 were measured at three temperatures 295 K, 77.4 K and 8.0 K. These spectra are plotted in Fig. 1. The isomer shift was evaluated by a least-squares fitting of Lorentzian shape to the data. In the case of the spectra obtained at 295 K and 77.4 K the experimental spectrum consist of a single resonance dip and it is therefore possible to fit the points by a single Lorentzian curve. The spectrum taken at 8.0 K is more complex, but the observed data are adequately described by the sum of two Lorentzians. The intensities of these two components are in a ratio close to 2:1 and their centers of gravity are at 0.50 mm/s and -1.53 mm/s, respectively.

The observed values of the isomer shift and the linewidth for Sm_3Se_4 , as well as those for Sm_2Se_3 and $\text{SmSe}^{6)}$, are collected in Table 1. In the latter two compounds the valencies of samarium are +3 and +2, respectively. Mössbauer absorption measurement in Sm_3Se_4 at 4.5 K is now in progress. Detailed results and discussions of valence fluctuating state on Sm_3Se_4 will be published later.

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Table 1. ^{149}Sm isomer shift and linewidth

Sample	Temperature	δ (mm/sec)	Γ (mm/sec)
Sm_3Se_4	295 K	-0.28	4.92
	77.4 K	-0.28	4.44
	8.0 K	+0.50	4.16
		-1.53	3.38
$\text{Sm}_2\text{Se}_3^{a)}$	Room Temp.	-0.02	2.56
$\text{SmSe}^{a)}$	Room Temp.	-0.71	2.15

a) Ref. 1.

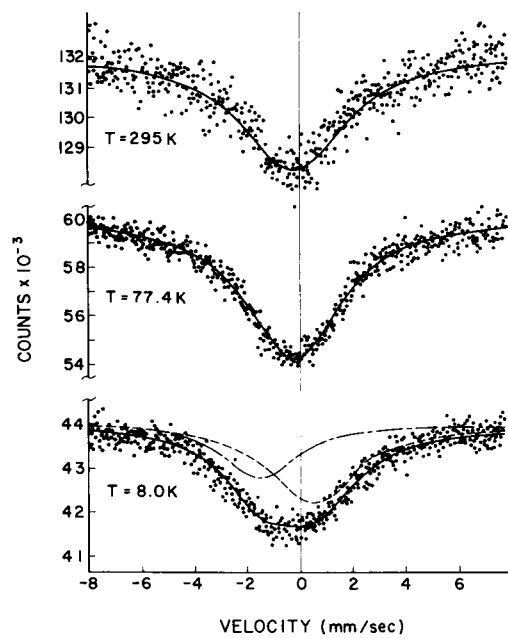


Fig. 1. Mössbauer spectra of Sm_3Se_4 . The solid line is a Lorentzian least-squares fit to the data.